Investigation of the decay property of extremely low-lying isomer ^{229m}Th

Y. Kasamatsu[1], H. Kikunaga[2], K. Takamiya[3], T. Mitsugashira[4], T. Nakanishi[2], T. Ohtsuki[4], H. Yuki[4], W. Sato[1], H. Yamana[3], Y. Ohkubo[3], H. Kimura[1], S. Shibata[3], Y. Kawase[3], A. Shinohara[1]

[1] Osaka University, Japan
[2] Kanazawa University, Japan
[3] Kyoto University, Japan
[4] Tohoku University, Japan

kasamatu@chem.sci.osaka-u.ac.jp

Introduction

The chemical and physical properties of the isomeric state of ^{229}Th, having the lowest excitation energy, is an interesting subject in both experiment and theory. Helmer and Reich reported that the excitation energy of ^{229m}Th is about 3.5 eV from the result of precise γ-ray spectroscopy for the α-decay of ^{233}U [1]. A simplified level scheme of ^{229}Th is shown in Fig.1. This level corresponds to a 3/2+ [631] Nilsson state while the ground state to a 5/2+ [633] one [2]. The emission of internal conversion electrons is forbidden because the excitation energy is lower than the first ionization energy of thorium atoms. Thus the deexcitation from ^{229m}Th to the ground state is expected to occur through a direct γ-ray transition. Furthermore, if the outer-shell electron of ^{229m}Th can be involved in the decay of ^{229m}Th nucleus, ^{229m}Th may decay via an electron bridge (EB) mechanism [3]. The diagram of EB mechanism is shown in Fig.2. This implies that the half-life of ^{229m}Th is dynamically variable depending on its chemical state. The photons emitted in a direct isomeric transition from this level to the ground state should have about 350 nm wavelength, and the photons involved in the transition via EB process are deduced to correspond to visible rays. However, there has been no direct observation for the transition of ^{229m}Th yet. The successful observation will allow us to research the details of EB mechanism.

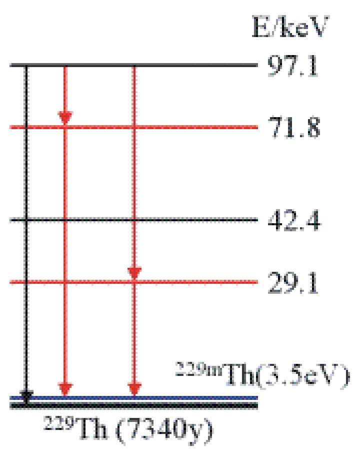

Figure 1. Level scheme of ^{229}Th.

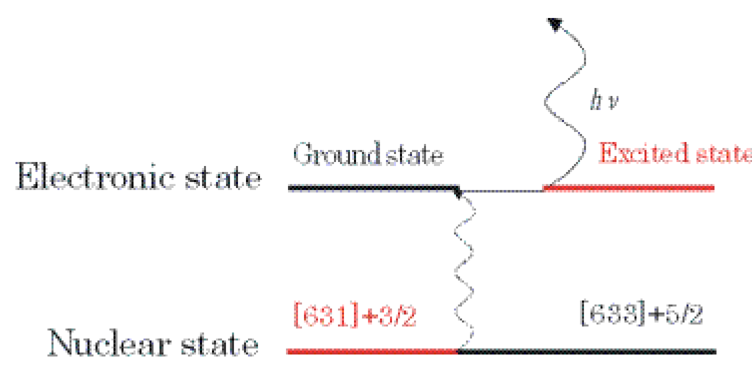

Figure 2. Diagram of Electron Bridge Mechanism.

The ^{233}U sample contains a given amount of ^{229m}Th produced through an α-decay from ^{233}U with a branching ratio of about 1-2 percents. Several kinds of experiments were performed by other groups for the observation of photons emitted from the ^{233}U sample [4,5]. These observations were not successful, however, owing to the α-particle-induced fluorescence of the materials (nitrogen and quartz, etc.) around the radioactive sample [6].

We introduce a new photon detection system and an α-ray spectroscopic technique to investigate the decay property of ^{229m}Th. The ^{229m}Th samples were prepared by two different methods, chemical separation of decay product from ^{233}U or ^{229}Ac, and direct production by several nuclear reactions. Here preliminary results are reported.

Experimental

The new method of production of ^{229m}Th was performed by novel method using several nuclear reactions: ^{228}Ra(n,γ)^{229}Ra, ^{232}Th(γ,p2n)^{229}Ac, ^{230}Th(γ,n)^{229}Th, ^{232}Th(p,p3n)^{229}Th and ^{230}Th(p,d)^{229}Th. ^{229}Ra decays into ^{229}Ac ($T_{1/2}$=~4m), and ^{229}Ac decays into ^{229}Th ($T_{1/2}$=62.7m). In the decay process from ^{229}Ac, ^{229m}Th is expected to be produced with high probability.

We tried to observe the decay from ^{229m}Th directly by two different methods to investigate the decay mechanism of ^{229m}Th. One is to detect the ultraviolet and visible photons from ^{229m}Th. Although this attempt has not been successful yet, it is essential to investigate the EB mechanism. Photon measurement was performed only for ^{229}Th samples separated from ^{233}U or ^{229}Ac, because the detector was very sensitive to each radiation and thermal phenomenon. The other method is to detect the α particle emitted from ^{229m}Th. We may observe the α rays from ^{229m}Th, since the partial half-life of α-decay in ^{229m}Th is considered to be shorter than that in ^{229}Th as will be mentioned below. The results obtained by the photon measurement can be attributed to the nuclear phenomenon by taking the results for the α-ray measurement into account.

1) Sample preparation

a) α-decay from ^{233}U

The half-life of ^{229m}Th has not been determined experimentally yet, estimated in wide range from about ~10^{-2}s to tens of hours [1,7]. Therefore we developed a rapid ion exchange apparatus so as to make successfully the measurement even when the lifetime was rather short.

The experimental procedure is as follows. First ^{233}U was adsorbed on an anion exchange resin layer in 8M hydrochloric acid solution. Making use of a chemical property that the daughter nuclides of ^{233}U cannot be adsorbed to the resin layer, ^{229}Th grown up during a certain time (Growth Time) was eluted and separated from ^{233}U. It takes only a few minutes for this separation.

Not only the elution peak but all the other range of the elution were also measured to confirm whether the photon emission derived from ^{229m}Th. The ^{229g}Th sample as well as the separated ^{229}Th sample was measured under the almost same conditions to evaluate the effects of fluorescence of thorium atoms and those of radiation from ^{229}Th nuclei.

b) ^{228}Ra(n,γ)^{229}Ra

^{228}Ra was prepared by separating Ra from a ^{232}Th sample in radio-equilibrium. The ^{232}Th sample (thorium nitrate $Th(NO_3)_4$-$5H_2O$) was dissolved in 0.5M Nitric acid. All elements in the decay-chain of ^{232}Th except Ra were precipitated by adding ammonia solution. After centrifuging Ra solution was obtained. Th was completely removed by anion exchange method. Then the ^{228}Ra solution sample was dried up on a quartz boat.

The neutron irradiation was performed in Kyoto University Reactor. ^{228}Ra of about 220kBq was prepared as a target. Neutron flux was 2.8×10^{13} n/s. The duration of irradiation was 60 minutes. After irradiation the sample was rapidly dissolved in 2M HCl, and Ac was separated by a cation exchange method. We measured the γ-ray energy to ascertain the production of ^{229}Ac. The separated Ac solution sample was divided in half, one was assayed for α-ray measurement. The other was left for 3hours until ^{229}Th was grown fully. Th was isolated by the anion exchange method, and a part of that was assayed for photon measurement and the residual for α-ray measurement.

c) ^{230}Th(γ,n)^{229}Th, ^{232}Th(γ,p2n)^{229}Ac

About 30 μg of 95% ^{230}Th molecular-plated on a 5N aluminum plate was enclosed in a quartz tube for Bremsstrahlung irradiation. About 1.5g of ^{232}Th oxide was also enclosed in a quartz tube and used as a target. The irradiation was carried out using the Electron Linear Accelerator of Tohoku University (Linac). The linac was operated at electron energies of 27 MeV with the beam pulse width of 3 μs the peak current around 100 mA, and the pulse repetition rate of 300 s^{-1}.

After the irradiation, thorium isotopes were chemically separated from the other nuclear reaction products and fission products by the anion exchange and cation exchange method. The thorium isotopes in the effluent were then coprecipitated with samarium trifluoride by adding 30 - 250 μg samarium and hydrofluoric acid solution.

d) ^{232}Th(p,p3n)^{229}Th, ^{230}Th(p,d)^{229}Th

Proton irradiation was carried out with AVF Cyclotron at Research Center for Nuclear Physics in Osaka University. 770 μg of ^{232}Th molecular-plated on a 5N aluminum plate was prepared as a target. Proton beam energy was 34-36 MeV, and the beam current was about 1 μA. The duration of irradiation was 8 hours.

About 5μg of ^{230}Th was used for irradiation. The target was exposed to about 14.8 MeV, 1 μA proton beam for 8 hours. Another target was irradiated for 1 hour in the same beam condition. Immediately after the irradiation, this sample was measured by silicon detector without chemistry to confirm the production of ^{228}Th, Pa and Ac. At this beam energy it is expected that the nuclear reaction producing compound nuclei would be inhibited by the coulomb barrier, only ^{229}Th should be produced.

Pa and fission products produced simultaneously were first removed by an anion exchange separation after the irradiation. After removal of aluminum by precipitation adding NaOH, the Th fraction was separated from Ac and other fractions by a cation exchange method. Acquired sample was coprecipitated with samarium, and assayed for an α-ray measurement.

2) Measurements

a) Photon measurement

Low noise photomultiplier (PM) was used for the photon detection. PM was installed in a PM cooler to lower the thermal noise. Further, the oval reflector was employed, as shown in Fig.3, to focus as many photons emitted from the sample as possible on the photocathode (5mm×8mm) of PM. The output signals from PM was transformed, through only Pre-Amplifier and Discriminator, to MCS-mode data collecting system.

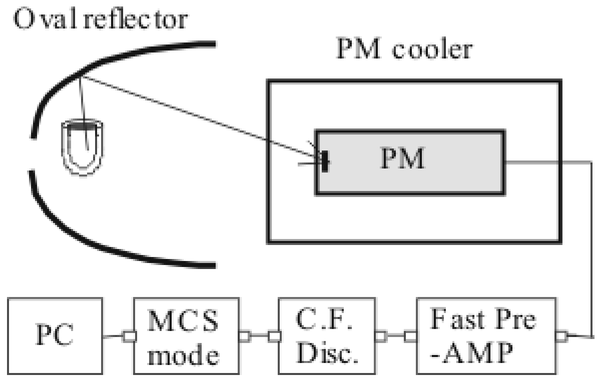

Figure 3. Photon detection system with oval reflector.

The sample measured was usually solution put in a quartz tube (ϕ8mm, 1cm). Several droplets of eluent at elution peak of thorium were collected in it.

b) α-ray measurement

The most favored α-transition from the ground state of ^{229}Th feeds to the 5/2+[633] state at 236.3 keV level of the daughter ^{225}Ra, and only rather weak α-transitions are observed in the higher energy range, as shown in the column for ^{229}Th in Table 1. The transition from ^{229}Th to the 149.96 keV 3/2+[631] level of ^{225}Ra (Eα=4.930 MeV) that is expected to be the most favored α-transition from ^{229m}Th has a branching ratio of only 0.16%. In addition, the α-transition to the parity coupled 3/2+ state at 42.77 keV that is assigned to the rotational band of the ground state of ^{225}Ra (1/2+[631]) is also expected to be another favored α-transition from ^{229m}Th. As a result, the α-particles from ^{229m}Th have higher energies than those from ^{229}Th. This implies that the partial half-life of the α-transition of ^{229m}Th is considerably shorter than that of ^{229}Th, and that the α-particle might be observable when ^{229m}Th is produced by a suitable reaction.

Table 1. Levels of ^{225}Ra and the α-transition ratio from ^{229}Th.

Orbit	Excitation Energy/keV	α-branch		Eα/Mev
		^{229}Th (%)	^{229m}Th	
[631]1/2+	0	weak	favored	5.079
3/2+	42.77	0.24	strong	5.036
5/2+	25.41	6.6	favored	5.053
7/2+	111.60	5.97		4.968
9/2+	100.5	3.17		4.978
[631]3/2+	149.96	0.16	strong	4.93
5/2+	179.75	10.2	weak	4.901
7/2+	243.6	5.0		4.836
[633]5/2+	236.3	56.2	weak	4.845
7/2+	267.9	9.3		4.815
9/2+	321.8	1.9		4.761

The sample for α-spectrometry was prepared by coprecipitating thorium isotope with samarium as fluoride or hydroxide on 0.1 μm or 0.02 μm pore size membrane filter. The precipitate was subjected to α-spectroscopy using a 450 or 900 mm^2 silicon detector.

Results and Discussion

1) Photon measurement

Photon emissions were observed for the solution samples separated from both ^{233}U and ^{229}Ac produced by ^{228}Ra(n,γ)^{229}Ra reaction. There was no decay component in the time dependence as shown in Fig.4. The eluent sample that is lying out of elution peak position also emitted a few visible or ultraviolet photons. It is difficult to attribute the origin of photon emission to ^{229m}Th nucleus.

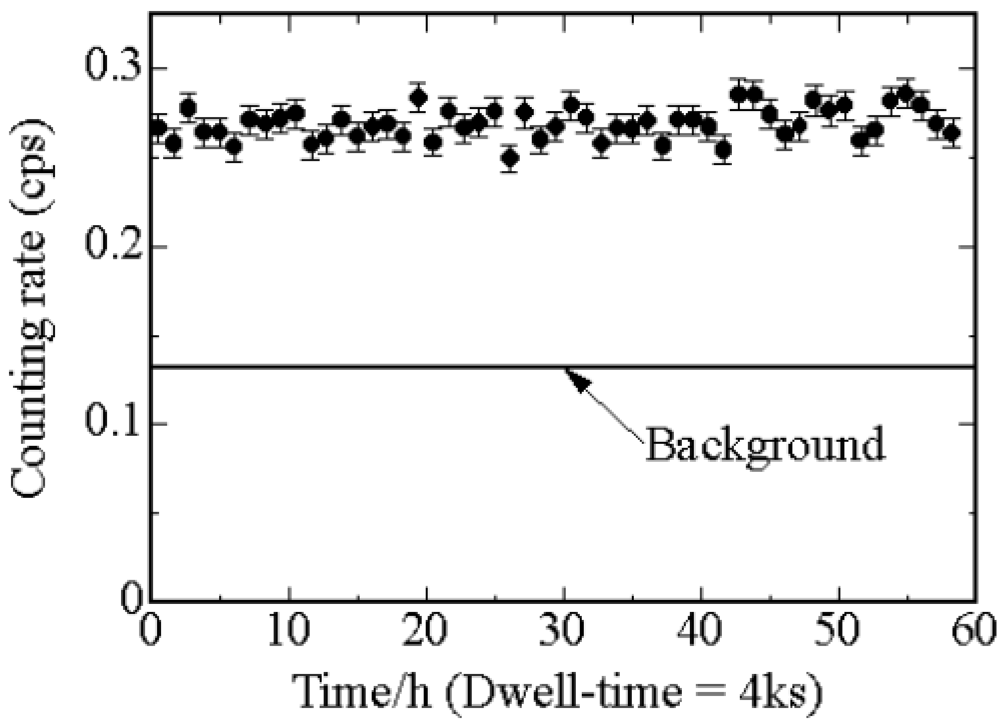

Figure 4. An exmaple of photon measurement result from ^{229}Th solution sample chemically separated from ^{233}U.

Little photon emission was observed from ^{229g}Th sample. The fluorescence of thorium atoms and the other photons derived from α-particle emission of ^{229g}Th do not contribute to the detected photons.

The transition energy of ^{229m}Th may lie out of the energy region of the present detector.

Development of a spectroscope for one-photon counting is under consideration to distinguish the decaying component of a certain wavelength. We are measuring photons with the detector working in higher energy region.

2) α-ray measurement

In the ^{228}Ra(n,γ)^{229}Ra experiment, α-ray spectrum in the region of interest for ^{229m}Th was obscure because of the disturbance of the tail of α-ray peaks of ^{228}Th. γ-rays from ^{229}Ac (164.5 keV) were measured and the production of ^{229}Ac was ascertained.

In the $^{230}Th(\gamma,n)^{229}Th$ and $^{232}Th(\gamma,p2n)^{229}Ac$ (see Fig.5) experiments, α-ray peak of ^{229g}Th appeared to be detected in the α-ray spectrum of both experiments. There were some peaks in the energy region of ^{229m}Th, but the contribution from ^{231}Pa disintegrated from ^{231}Th was not negligible for the experiment using ^{230}Th target. The half-life of ^{229m}Th would be too long as compared with the estimation, if the α-decay of ^{229m}Th were observed as well as ^{229g}Th.

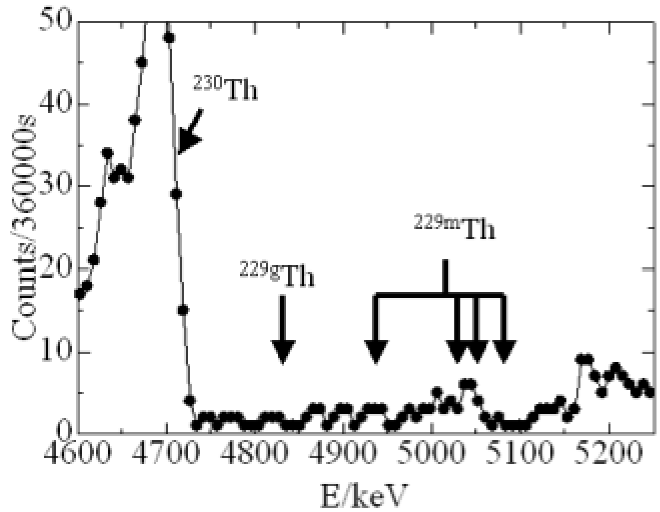

Figure 5. α-ray measurement result of ^{229}Ac sample chemically purified from $^{232}Th(\gamma,p2n)^{229}Ac$ reaction products.

In the $^{232}Th(p,p3n)^{229}Th$ (see Fig.6) and $^{230}Th(p,d)^{229}Th$ experiments, α-rays from ^{229g}Th were observed. However, these of ^{229m}Th were not measured clearly.This result implies that the half-life may be shorter than a few hours.

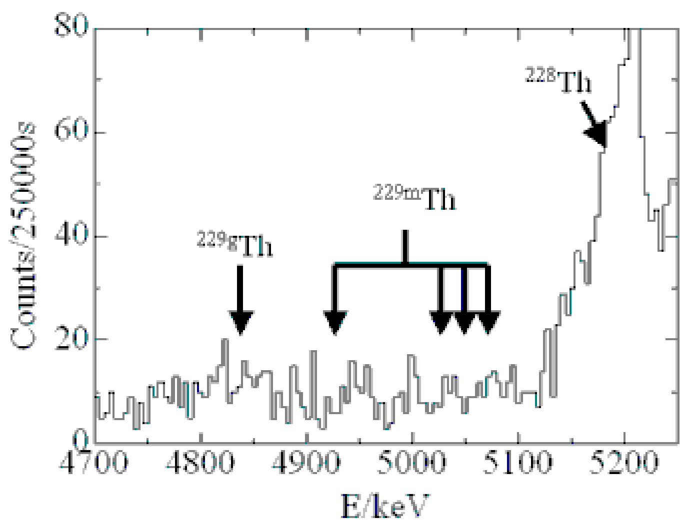

Figure 6. α-ray measurement result of ^{229}Th sample chemically separated from $^{232}Th(p,p3n)^{229}Th$ reaction products.

Consequently we could not observe the decay of ^{229m}Th or determine the half-life from the present results. Additional experiments and improvement of the detection methods are in progress.

References

[1] R. G. Helmer and C. W. Reich, Phys. Rev. **C49**, 1845(1994).
[2] D. G. Burke, P. E. Garrett, and T. Qu, Phys. Rev. **C42**,499 (1990).
[3] V. F. Strizhov and E. V. Tkalya, Sov. Phys. JETP **72**, 387(1991).
[4] D. S. Richardson *et al*., Phys. Rev. Lett. **80**, 3206 (1998).
[5] R. W. Shaw, J. P. Young, S. P. Cooper and O. F. Webb, Phys. Rev. Lett. **82**, 1109 (1999).
[6] J. P. Young, R. W. Shaw and Oren F. Webb, Inorg. Chem. **38**, 5192 (1999).
[7] A. M. Dykhne and E. V. Tkalya, JETP Lett. **67**, 251 (1998).